Electronic Structure of $LaRu_2Si_2$

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INTRODUCTION

A common research tool in the study of highly-correlated *f*-electron systems is the systematic comparison, both theoretically and experimentally, of the bulk properties for similar systems with only the *f*-occupation being varied. In this manner, the *f*-correlation and hybridization effects between localized *f*-orbitals and other band-like *s-p-d* states can be elucidated. The XRu_2Si_2 system with X=(La, Ce, Th, U) is a favorable isostructural series for characterization with the Ce $4f$ or U $5f$ occupation being varied from the non-*f* La and Th compounds. The focus of this work is the electronic structure and Fermi surface (FS) topology of the $4f^0$ rare-earth system of $LaRu_2Si_2$ as measured by angle-resolved photoemission (ARPES). Detailed comparison to LDA band structure calculations provides the framework for subsequent analysis of the $4f^1$ $CeRu_2Si_2$ system for which systematic differences in FS topology are predicted by LDA [1, 2]. $CeRu_2Si_2$ is also a prime literature example of good agreement between renormalized LDA calculations and de Haas van Alphen (dHvA) FS and effective mass measurements [3].

EXPERIMENT

The XRu_2Si_2 systems have the $ThCr_2Si_2$ crystal structure with a body-centered tetragonal Brillouin zone (BZ). A single crystal $LaRu_2Si_2$ sample was cleaved in ultra-high vacuum ($<7\times10^{-11}$ torr) at <135 K exposing the [001] surface. ARPES measurements were performed at ALS Beamline 7.0.1.2 with a total instrumental resolution of ≈80 meV and full angular acceptance of ≈0.7°. The Fermi-edge intensity mapping technique using automated angular motions was employed to gain an overview of the sample orientation and Fermi surface structure . Measurement at multiple photon energies corresponding to high symmetry ***k***-points at normal emission was used to explore k_z-dependences resulting from the three-dimensional BZ. Valence spectra were then acquired along the [100] and [110] high symmetry azimuth directions for comparison to LDA band structure calculations.

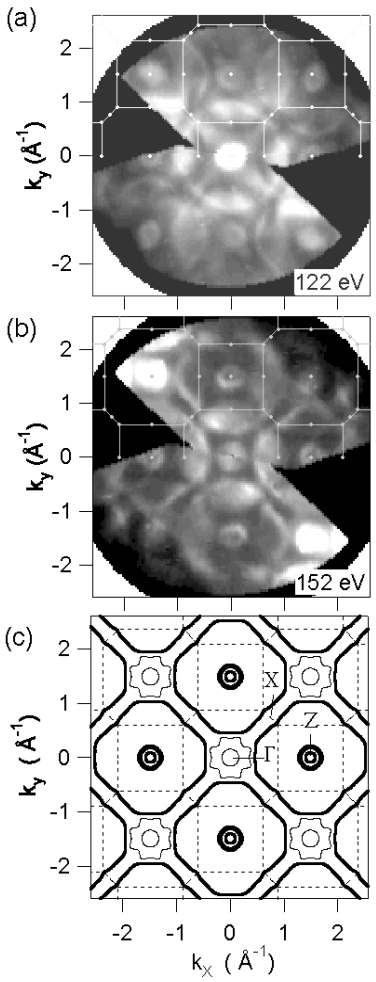
RESULTS

Fig. 1(a,b) shows E_F-intensity maps of $LaRu_2Si_2$ acquired at hν=122 eV and 152 eV which correspond to the Z and Γ points, respectively, at normal emission. The maps were acquired over a 120° azimuth and a 30° polar angle range, and have been corrected for misorientation of the sample surface to the sample goniometer rotation axes, two-fold symmetrized, and plotted as a projection onto the k_x-k_y plane. BCT Brillouin zone borders are overplotted in the upper half of the maps using the corresponding normal emission Z and Γ ***k***-points. Three main pieces of FS are observed to repeat in multiple Brillouin zones in the experimental maps of Fig. 1(a,b): *(i,ii)* small circular closed sheets centered at Γ and Z and *(iii)* a large sheet centered at Z that extends into the second BZ. The two maps in Fig. 1 do not distinguish the disconnectedness along k_z of the small FS pockets at Γ and Z. In fact, since Γ and Z alternately repeat along vertical k_z lines, the data could be interpreted as showing a single piece of FS that has been broadened along k_z. However, strong evidence for three-dimensionality of the FS and the existence of experimental k_z -resolution is provided by the large FS contour which is observed to shift from a Z-point center at normal emission at 122 eV to Z-point centers in the outer BZ's at 152 eV, consistent with the stacking of

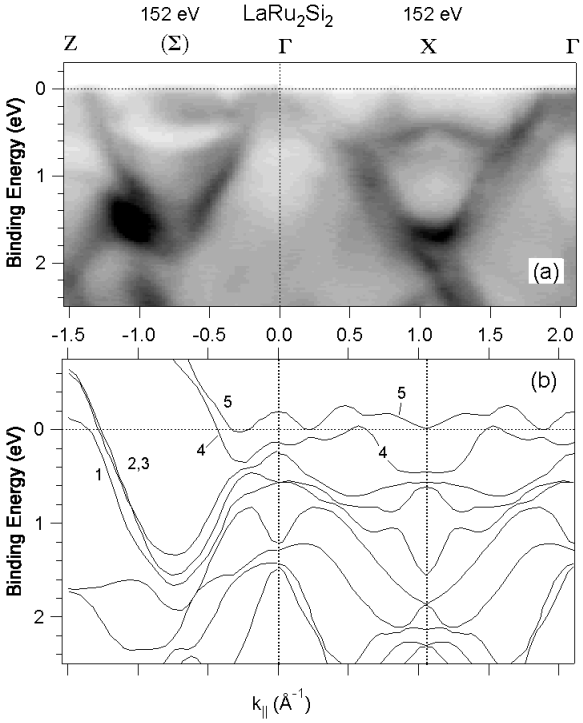
bct Brillouin zones in ***k***-space. Similar but fainter large FS contours centered around Γ are also observable and may result from k_z-broadening and/or the hemispherical curvature of the constant energy surfaces being measured in ***k***-space.

A schematic of the theoretical FS contours [1] is shown in Fig. 1(c) where bold (fine) lines indicate theoretical hole (electron) Fermi surfaces. Band theory predicts: *(i)* three concentric hole pockets centered on Z, *(ii)* very small electron pockets offset from and surrounding Γ, and *(iii)* a large hole surface centered on Z whose diameter extends into the next BZ's. The qualitative agreement between the experimental maps and the theory is very good, although the experimental FS maps do not resolve the small Z point contour into three concentric pockets, nor do they distinguish the electron or hole character of the FS contours, which can be determined only from valence spectra dispersions near E_F.

Fig. 2(a) shows reverse grayscale images of $LaRu_2Si_2$ valence band spectra acquired at hν=152 eV with 0.5° polar angle steps along two azimuth angles separated by 45° that correspond to the Γ–(Σ)–Z and Γ–X–Γ high symmetry directions. For comparison, the theoretical band structure along the same two directions is shown in Fig. 2(b). Ignoring the weaker features in the experimental data and various detailed discrepancies with theory, major overall agreement with theory can be identified. *(i)* The Z point FS is verified experimentally to be a hole pocket that originates from parabolic bands that disperse symmetrically along Z–(Σ)–Γ.



(left) Figure 1. Fermi-energy intensity maps of $LaRu_2Si_2$ at (a) 122 eV and (b) 152 eV. (c) Theoretical contours of hole (bold) and electron (fine) Fermi surface topologies.



(above) Figure 2. (a) Valence band intensity maps of $LaRu_2Si_2$ at 152 eV along high-symmetry azimuth angles corresponding to Γ–(Σ)–Z and Γ–X–Γ. (b) Theoretical band structure calculation for $LaRu_2Si_2$.

(ii) Band theory shows a complicated region near Γ arising from band hybridizations. A key feature, though, is the existence of a mostly unoccupied band (5) just above E_F at Γ that dips below E_F forming tiny electron pockets at $k_x \approx -0.25$ Å^{-1}. The existence of such an electron pocket is readily apparent in the experimental data along Γ–(Σ)–Z, centered at $k_x = -0.35$ Å^{-1} with 0.3 eV band minimum. However, the outer edge of the experimental electron pocket corresponds to the outer-edge of the large contour in the FS maps of Fig. 1, i.e. that has the connectivity of the large hole-surface of band 4. A possible reconciliation of this discrepancy is that band 5 does not actually cross E_F while band 4 recrosses E_F before the Γ-point. This interpretation is not contradicted by dHvA data, which also are not in detailed agreement with LDA in this region [1], and agrees better with the valence band image in Fig. 2(a), which shows that bands (1-3) disperse very close to E_F at Γ, possibly even forming a small hole surface.

(iii) Along Γ–X–Γ we observe a dominant band dispersion that is symmetric about the X point also with a band minimum of ≈1.6 eV and a second band dispersing downwards from the X-point at ≈ 0.5 eV binding energy. Both of these features are present in the band theory, albeit with a greater splitting of the bands. The band theory also predicts small electron pockets (above and below) the X-point (visible only with 3D FS figures) originating from fairly flat dispersions that are probably very sensitive in size to band dispersions and location of the chemical potential. Some experimental weight at E_F near the X-point has the wrong dispersion compared to the theoretical bands, but may be related to the hypothesis concerning bands 4 and 5 in the preceding paragraph.

CONCLUSION

Hence we have established the ability of ARPES to identify the basic band structure and FS topology of $LaRu_2Si_2$, and have demonstrated the basic validity of the LDA band structure calculations. Good agreement between experiment and theory is observed not only below 0.5 eV binding energy, but also in the FS region including large hole surfaces centered on Z in $LaRu_2Si_2$ and small electron pockets around Γ. The large hole surface centered on Z is the important "pillow" FS which is predicted to become very heavy in $CeRu_2Si_2$ [4]. Concurrent research builds on this agreement and understanding of the $4f^0$ system to study and identify correlation effects in the $4f^1$ system of $CeRu_2Si_2$ [5,6].

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This work was supported at U. of Michigan by the U.S. Dept. of Energy (DoE) under contract No. DE-FG02-90ER45416 and by the U.S. NSF under grant No. DMR-99-71611; and at LANL by the U.S. DoE.

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